Nature of weak magnetism in $SrTiO_3/LaAlO_3$ multilayers

Z. Salman,[1,*] O. Ofer,[2] M. Radovic,[3,4] H. Hao,[5] K. H. Chow,[6] M. D. Hossain,[5] C. D. P. Levy,[2] W. A. MacFarlane,[7] G. M. Morris,[2] L. Patthey,[3] M. R. Pearson,[2] H. Saadaoui,[1] T. Schmitt,[3] D. Wang,[5] and R. F. Kiefl[5,2]

[1]*Laboratory for Muon Spin Spectroscopy, Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland*
[2]*TRIUMF, 4004 Wesbrook Mall, Vancouver, BC, Canada, V6T 2A3*
[3]*Swiss Light Source, Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland*
[4]*Laboratory for Synchrotron and Neutron Spectroscopy, Ecole Polytechnique Federale de Lausanne, CH-1015 Lausanne, Switzerland*
[5]*Department of Physics and Astronomy, University of British Columbia, Vancouver, BC, Canada V6T 1Z1*
[6]*Department of Physics, University of Alberta, Edmonton, AB, Canada T6G 2E1*
[7]*Department of Chemistry, University of British Columbia, Vancouver, BC, Canada V6T 1Z1*

We report the observation of weak magnetism in superlattices of $LaAlO_3/SrTiO_3$ using β-detected nuclear magnetic resonance. The spin lattice relaxation rate of ^{8}Li in superlattices with a spacer layers of 8 and 6 unit cells of $LaAlO_3$ exhibits a strong peak near ~ 35 K, whereas no such peak is observed in a superlattice with spacer layer thickness of 3 unit cells. We attribute the observed temperature dependence to slowing down of weakly coupled electronic moments at the $LaAlO_3/SrTiO_3$ interface. These results show that the magnetism at the interface depends strongly on the thickness of the spacer layer, and that a minimal thickness of $\sim 4-6$ unit cells is required for the appearance of magnetism. A simple model is used to determine that the observed relaxation is due to small fluctuating moments ($\sim 0.002\ \mu_B$) in the two samples with a larger $LaAlO_3$ spacer thickness.

The electronic, magnetic and structural properties of an interface between two materials is in general different from the bulk properties of both. A dramatic example, discovered recently [1–3], is the high mobility two-dimensional electron gas (2DEG) at the interface between two insulating perovskite oxides; TiO_2-terminated $SrTiO_3$ (STO) and $LaAlO_3$ (LAO). Surprisingly, there is evidence that this interface can be both magnetic[4, 5] and even superconducting below ~ 300 mK[6]. It is generally agreed that these properties are associated with subtle structural changes at the interface. Several attempts have been made to explain the high carrier densities at the interface, including doping with electrons or oxygen vacancies [2, 7–10], inter-diffusion [9, 11, 12], and the influence of lattice distortions [13–17, 33]. However, the details and mechanism behind the observed properties seem to involve several processes [10–12, 18–21].

The unusal properties of the LAO/STO interface is extremely relevant for the general interface phenomena at oxide and perovskite interfaces [22, 23]. This important class of materials exhibits a variety of physical properties including magnetic [24–27], superconducting [6], insulating and conducting [1, 6, 24, 28]. The observed weak magnetism in this particular LAO/STO system may have significant implications on the interpretation of interface properties and proximity effects in other oxides. However, since both materials in this case are non-magnetic and insulating the appearance of weak magnetism and conductivity can be easily detected.

In this Letter we address questions concerning the nature of the reported magnetism at the interfaces between LAO and STO. To date, most reports of magnetism at these interfaces are indirect being based on transport measurements at high applied magnetic field and limited to bi-layers. More recent reports have contradicting claims of coexistence [29, 30] and phase separation [31] of superconductivity and magnetism. These studies report measurements on bi-layers of LAO on TiO_2 terminated STO, and currently more efforts are being invested in producing superlattices (SLs) of LAO/STO[32, 33]. This is to address the question whether the interfaces in SLs maintain the same properties as bi-layers, but also to answer the question whether the TiO_2 termination of the STO substrate, and the subsequent polar ctastrophe scenario, is crucial in this case [1, 19]. Recent polarized neutron reflectometry (PNR) has concluded there is no detectable magnetism in SLs, putting a very small upper limit on any possible magnetization [32]. In fact, until now there has been no direct observation of internal magnetic fields (in either bi-layers or SLs) that must be present in any true magnetic state. Here we report such results using β-detected nuclear magnetic resonance (β-NMR) measurements in SLs of LAO/STO. For SLs with a LAO spacer layer exceeding a "critical" thickness, we find the spin lattice relaxation rate of polarized ^{8}Li exhibits a strong temperature dependence with a maximum at $T^* \sim 35$ K. This behaviour is typical of a slowly fluctuating internal magnetic field expected near a magnetic transition at T^*, and provides direct evidence of magnetism at the interface between insulating and nonmagnetic LAO and STO. The weak magnetism is attributed to localized charge carriers at the interface. We estimate that the size of the magnetic moment per unit cell (uc) is about $\sim 1.8 \times 10^{-3}\ \mu_B$, indicating the moments are only weakly dependent of the LAO spacer thickness beyond a critical value of $4-6$ uc.

The β-NMR technique is a magnetic resonance technique similar to both nuclear magnetic resonance and

muon spin relaxation (μSR). The local spin probe used here is ^{8}Li. A low energy (28 keV) beam of radioactive ^{8}Li is produced at the isotope separator and accelerator (ISAC) at TRIUMF in Vancouver, Canada. It is then spin-polarized using a collinear optical pumping method, yielding nuclear polarization in excess of 70%, and subsequently implanted into the sample. Since the implanted beam energy can be adjusted, the ^{8}Li mean stopping depth can be varied between 1-250 nm. The nuclear polarization, and its time evolution, is the quantity of interest in these experiments. It can be measured through the β-decay asymmetry, where an electron is emitted preferentially opposite to the direction of the nuclear polarization at the time of decay [34] and detected by appropriately positioned scintillation counters. ^{8}Li is a spin $I = 2$ nucleus with a small electric quadrupole moment $Q = +31$ mB and gyromagnetic ratio $\gamma = 6.301$ MHz/T. The spin lattice relaxation of the ^{8}Li nuclear spin can be measured by implanting a short pulse of beam for a duration t_p (e.g. 1 second), and measuring the polarization as a function of time, $p_z(t)$, during and after the beam pulse. More details about the techniques can be found in Refs. [35–37].

Measurements on three different SLs are reported here. These were grown using pulsed laser deposition and consist of 10 LAO/STO stacking periods grown on TiO_2 terminated $\langle 100\rangle$ single crystal STO substrates. The thickness of the LAO layers were $n = 8$, 6 and 3 uc, while the STO layers are fixed at 10 uc [33]. Hereafter, we refer to these SLs as LAOn, where n is the number of uc in the LAO layers. After growth, the samples were annealed for 5 hours at 1000°C in 1 bar of O_2 in order to fill oxygen vacancies [33]. All LAOn samples were investigated using resonant inelastic X-ray scattering (RIXS), and their preparation details are given in Ref. [33]. Additional control measurements were also performed on STO and LAO single crystals obtained from Crystec GmBH.

Typical relaxation curves measured in LAO8 and LAO3, using 5 keV ^{8}Li implantation energy, are shown in Fig. 1(a) and (b), respectively. This implantation energy corresponds to mean implantation depth of ~ 20 nm in the samples. In these measurements $p_z(t)$ is determined by both the ^{8}Li spin-lattice relaxation rate, $\lambda = 1/T_1$, and its radioactive lifetime, $\tau = 1.21$s. Assuming a beam pulse duration t_p and a general spin relaxation function $f(t, t' : \lambda)$ for the fraction of ^{8}Li implanted in the sample at t', the polarization follows [36]

$$p_z(t) = \begin{cases} \dfrac{\int_0^t e^{-(t-t')/\tau} f(t,t':\lambda)dt'}{\int_0^t e^{-t/\tau}dt} & t \le t_p \\ \dfrac{\int_0^{t_p} e^{-(t_p-t')/\tau} f(t,t':\lambda)dt'}{\int_0^{t_p} e^{-t/\tau}dt} & t > t_p. \end{cases} \quad (1)$$

The data in Fig. 1 are best fit to Eq. (1) with a phenomenological stretched-exponential form,

$$f(t, t' : \lambda) = Ae^{-[\lambda(t-t')]^{0.3}}. \quad (2)$$

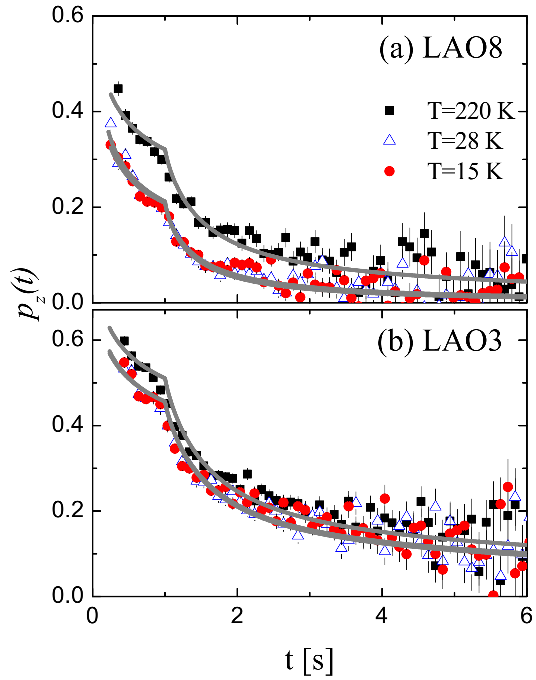

FIG. 1: (Color online) ^{8}Li spin relaxation curves measured in (a) LAO8 and (b) LAO3 in 3 mT applied field, 5 keV ^{8}Li implantation energy and various temperatures. Note the relaxation rate is larger and more temperature dependent in LAO8 compared to LAO3. Note also the long lived tail in LAO3 which is absent in LAO8.

A much stronger temperature dependence is observed in both LAO8 and LAO6, with a relaxation rate which is generally higher than that observed in LAO3. In Fig. 2 we plot the relaxation rates in all SLs as a function of

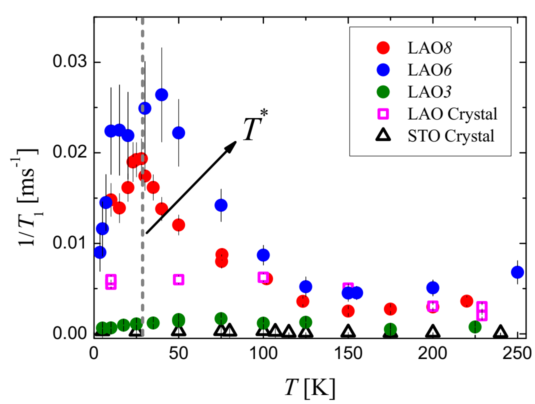

FIG. 2: (Color online) The spin lattice relaxation rate ($1/T_1$) as a function of temperature in 3 mT applied field. The red, blue and green circles are measurements in LAO8, LAO6 and LAO3, respectively. The squares and triangles are reference measurements in LAO and STO bare crystals.

temperature compared to the relaxation rates measured in single crystals of LAO and STO under the same conditions. As expected, the relaxation in STO is much

smaller than that measured in LAO, where the fluctuating Al nuclear moments contribute to the relaxation of the ^{8}Li spin at these low fields [36, 38]. Note, in both STO and LAO single crystals, there is only weak temperature dependence. LAO3 exhibits a similar weak temperature dependence, while LAO8 and LAO6 show a clear increase in $1/T_1$ as the temperature is lowered, followed by a pronounced peak near $T^* \sim 35$ K and a decrease as the temperature is lowered further. Note, the strong temperature dependence and enhancement in $1/T_1$ at low temperatures in LAO6 and LAO8 is not present in intrinsic LAO or STO. We attribute this behaviour to slow magnetic fluctuations, due to magnetic freezing or critical slowing down of magnetic fluctuations at the interfaces occurring near T^*.

The relaxation in LAO8 and LAO6 at high temperature approaches that measured in bulk LAO, while in LAO3 the relaxation at high temperature is somewhere between that of LAO and STO. This difference can be understood as an average contribution of LAO and STO layers in the different SLs. At the 5 keV ^{8}Li implantation energy we estimate that the ratio between ^{8}Li stopped in STO:LAO is $\sim 1.3:1$ in LAO8, $\sim 1.7:1$ in LAO6, and $\sim 3.4:1$ in LAO3. Therefore, our results indicate that at high temperatures the relaxation in the SLs is dominated by fluctuating nuclear moments in LAO (though some contribution of the magnetic fluctuations are still present in LAO8 and LAO6). However, at lower temperatures there is clear evidence of a different relaxation mechanism developing in LAO8 and LAO6, which is not present in LAO3 or intrinsic STO or LAO.

Recent RIXS measurements on the same samples have revealed localized as well as delocalized Ti 3d carriers in such SLs [33]. These were attributed to spin-bearing Ti^{3+} ions at the interface. An orthorhombic structural distortion of $Ti^{3+}O_6$ octahedra was also observed. However, while the density of charge carriers depends on the thickness of the LAO layers, n, the distortion of the $Ti^{3+}O_6$ does not. Furthermore, the annealing process was found to reduce significantly the density of both types of carriers due to the reduction of Ti^{3+} to Ti^{4+}[39], but it does not affect the orthorhombic distortion at the interfaces. From these measurements it was concluded that, for the annealed SLs, there is a critical thickness of 6 LAO uc, above which the density of carriers increases dramatically [33].

Our spin lattice relaxation measurements demonstrate that the LAO8 and LAO6 samples exhibit significantly enhanced spin relaxation at low temperatures compared with LAO3. More importantly, we see a distinct anomaly near T^*, possibly related to the onset of the static magnetism reported near 35 K [5]. A priori, the peak at T^* could have a non-magnetic origin. For example, temperature dependent fluctuations in the electric field gradient (EFG) at the ^{8}Li site, which couple to its electric quadrupole moment [36] (e.g. a ferroelectric transition). However, we can rule out EFG fluctuations since (I) RIXS measurements confirm that the non-cubic distortions in these SLs do not depend on the thickness of the LAO layers (and so do their contributions to $1/T_1$), and (II) we do not observe a strong temperature dependence in LAO3. Hence, the $1/T_1$ enhancement in LAO8 and LAO6 must have a magnetic origin, and therefore, almost certainly due to localized charge carriers at the interface. In what follows, we evaluate the average size of the magnetic moments per unit cell, assuming that the magnetism is concentrated at the LAO/STO interfaces.

The ^{8}Li probes are implanted almost uniformly within the volume of the SLs. Using our $1/T_1$ results in the magnetic SLs we can estimate the size of fluctuating local magnetic fields, Δ, experienced by the ^{8}Li. In the fast fluctuation limit we can write [40],

$$\frac{1}{T_1} = \frac{\gamma^2 \Delta^2 \tau_c}{1+\omega^2\tau_c^2}, \tag{3}$$

where τ_c is the correlation time of magnetic field fluctuations and ω is the precession frequency of the spin probe. In the presence of strong quadrupolar interactions, as in STO and LAO, ω is dominated by the quadrupolar frequency of the transition $m = \pm 2 \rightarrow \pm 1$. This can be estimated at ~ 230 kHz in STO [35]. We assume for simplicity that the maximum in $1/T_1$ corresponds to a T_1 minimum such that τ_c satisfies $\omega\tau_c \sim 1$ [40]. In this case, we can estimate $\Delta \simeq 4.8 \times 10^{-4}$ and 5.4×10^{-4} T for the LAO8 and LAO6, respectively.

One can also estimate the size of the moment needed to produce such magnetic fields using a few simplifying assumptions. First we assume there is a lattice of magnetic moments, $\mu = \alpha\mu_B$ (α is a constant and μ_B is the Bohr magneton), arranged on a square lattice (a) at the interfaces. We then calculate the distribution of dipolar fields experienced by a ^{8}Li, located at a distance z from the interface, by summing up the contributions from all moments [37, 41] (see schematic in Fig. 3). The root mean square (RMS) of the distribution falls as $\sim 1/z^2$ away from the interface [37, 41]. Therefore, the resulting RMS averaged over all implanted ^{8}Li (assuming a uniform distribution within all layers) is,

$$\Delta_{\mathrm{th}} \simeq C_0 \frac{\alpha}{a^3}, \tag{4}$$

where C_0 is a parameter that depends on the LAO layer thickness, a is in units of Å, and the resulting Δ_{th} is in Tesla. From these calculations we find $C_0 = 17.36$ for LAO8 and 18.95 for LAO6. Taking $a \sim 4$ Å as the unit cell of LAO (and STO) we find that $\Delta_{\mathrm{th}} \simeq 0.271\alpha$ T and 0.296α T for LAO8 and LAO6, respectively. Note that for $\mu = 1\mu_B$, Δ_{th} is about two to three orders of magnitude larger than the Δ estimated from $1/T_1$. Thus, our measurements imply an average magnetic moment of $\sim 1.8 \times 10^{-3}$ μ_B per unit cell at the LAO/STO interfaces, *equal in both* LAO8 and LAO6 samples. The

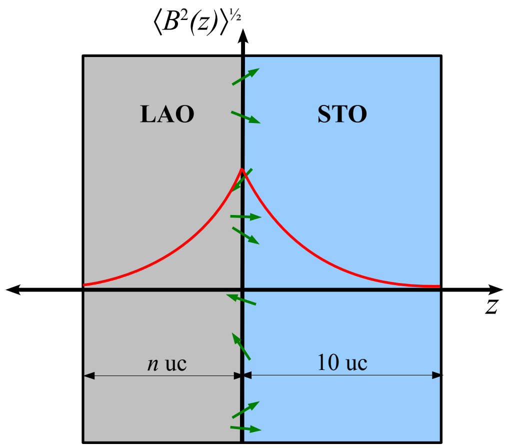

FIG. 3: (Color online) A schematic of magnetic moments (green arrows) at the LAO/STO interface. The red lines represent are the RMS of dipolar field distribution experienced by the ^{8}Li.

difference in $1/T_1$ is simply due to the different thickness of LAO layers. This result is consistent with (and confirms) the assumption that in the magnetic SLs the moments are confined to the interfaces and further indicates that their average size is independent of the LAO spacer layer thickness beyond the critical value. The small magnetic moment also explains why it has been missed with less sensitive techniques such as PNR [32].

It is likely that the observed magnetization is not uniformly distributed over the interface. If instead, we assume that there is an inhomogeneous distribution of $1\mu_B$ moments, then our calculations imply a two-dimensional spin density of $\sim 1.13 \times 10^{12}$ μ_B/cm^2. Surprisingly, this is of the same order of estimates from scanning SQUID measurements in bi-layers, $\sim 7.3 \pm 3.4 \times 10^{12}$ μ_B/cm^2 [29]. The small difference could be simply due to a different sample preparation procedure or a difference between bi-layers and superlattices. Moreover, there is a fundamental difference between the two. In a bi-layer the interface is formed between a TiO_2 terminated STO and a LaO^+ terminated layer of LAO, i.e. TiO_2/LaO^+. In contrast, it can be either TiO_2/LaO^+ or SrO/AlO_2^- in the SLs. These two types of interfaces have dramatically different electronic properties [1], in relation to the polar catastrophe [19] due to the different net charge of the LaO^+ and AlO_2^- layers. It is important to point out here that our results are consistent with the magnetism residing on *both types* of interfaces. Finally, the broad $1/T_1$ peak (in temperature) in the magnetic samples is further indication of the dilute and disordered magnetic moments at these interfaces (typically seen in dilute spin glasses [42]), in agreement with Ref. [29].

In conclusion, β-NMR of low energy ^{8}Li was used to investigate SLs of LAO/STO. We present direct evidence for weak magnetism in these SLs, attributed to a dilute concentration of magnetic moments at the interfaces. Our measurements agree with previous reports of this phenomenon in bi-layers of LAO/STO [4, 5, 29–31], but exhibit a surprising dependence on the thickness on the LAO layers. The magnetism is observed only in SLs with LAO layers exceeding a "critical" thickness of $4-6$ uc. This provides strong evidence for a direct connection between the observed magnetism and localized charge carriers detected in RIXS [33]. Furthermore, we find that the magnetism seems to be highly disordered and displays evidence of critical slowing down and possibly freezing near $T^* \sim 35$ K. A simple model calculation shows that it can be attributed to a two-dimensional spin density of localized magnetic moments of $\sim 1.13 \times 10^{12}$ μ_B/cm^2 which is independent of the thickness of LAO layers in magnetic SLs. This value is slightly lower than that found in bi-layers [29], nevertheless, it could explain its absence in the PNR data [32], since it does not produce sufficient contrast between the opposite neutron polarizations. Furthermore, our results demonstrate that, unlike the 2DEG, the magnetism apears on both types of STO/LAO interfaces, and therefore is unrelated to the polar catastrophe scenario. This indicates that the mechanism behind the 2DEG and magnetism may be different. Finally, our results establish a very stringent test for any robust theory attempting to explain the observed phenomena at the LAO/STO interfaces. We also note that these results may have significant implications on the interpretation of interface phenomena in oxide and perovsike materials in general [22, 23].

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* Electronic address: zaher.salman@psi.ch

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